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14. ABSTRACT

The possibilities of compensating losses in negative index metamaterials (NIMs) are shown without the requirements of population inversion. The approach relies on coherent nonlinear-optical energy transfer from the control optical field(s) to the NI signal through three- (TWM) and four-wave mixing (FWM) propagation processes. The feasibilities of tailoring the transparency of plasmonic slabs from strongly opaque through fully transparent to amplifying states with the control laser(s) as well as the possibilities for unparallel applications of the

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Compensating losses in optical negative-index materials and new frontiers in nanophotonics (final progress report)

ABSTRACT

The possibilities of compensating losses in negative index metamaterials (NIMs) are shown without the requirements of population inversion. The approach relies on coherent nonlinear-optical energy transfer from the control optical field(s) to the NI signal through three-(TWM) and four-wave mixing (FWM) propagation processes. The feasibilities of tailoring the transparency of plasmonic slabs from strongly opaque through fully transparent to amplifying states with the control laser(s) as well as the possibilities for unparallel applications of the discovered unique propagation properties are demonstrated. Two options are explored. One concerns the possibilities associated with the TWM nonlinearities intrinsic to the building blocks of the NIMs. The other one is aimed at independent engineering of resonantly enhanced FWM nonlinearities and NI. FWM nonlinearities are introduced by doping the host NIM with the resonant centers tailored by means of quantum control. The possibilities for creation of ultraminiature narrow-band frequency-tunable filters, reflectors, switches and cavityless generators of counter-propagating entangled right- and left- handed photons have been proved through numerical experiments.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

1. A. K. Popov, S. A. Myslivets and V.M. Shalaev, "Plasmonics: nonlinear optics, negative phase and transformable transparency (invited paper)," Plasmonics: Nanoimaging, Nanofabrication, and their Applications V, edited by Satoshi Kawata, Vladimir M. Shalaev, Din Ping Tsai, Proc. of SPIE Vol. 7395, 73950Z-1(12).

2. A. K. Popov, S. A. Myslivets and V.M. Shalaev, "Coherent nonlinear optics and quantum control in negative-index metamaterials," J. Opt. A: Pure Appl. Opt. 11, 114028-13 (2009).

3. A. K. Popov, S. A. Myslivets and V.M. Shalaev, "Coherent Nonlinear-optical Energy Transfer and Backward-wave Optical Parametric Generation in Negative-index Metamaterials," Physica B, (2010) doi:10.1016/j.physb.2010.01.022 (Published online January 6, 2010).

4. A. K. Popov and S. A. Myslivets, "Numerical Simulations of Negative-Index Nanocomposite and Backward-Wave Photonic Microdevices," World Academy of Science, Engineering and Technology, v.61, 107-121, January (2010), http://www.waset.org/journals/waset/v61/v61-16.pdf .

5. A. K. Popov, "Nonlinear optics of backward waves and extraordinary features of plasmonic nonlinear-optical microdevices," Eur. Phys. J. D (2010),
DOI: 10.1140/epjd/e2010-00097-4 (Published online April 20, 2010).

6. A. K. Popov and S. A. Myslivets, "Nonlinear-optical metamirror," Applied Physics A (Springer), Special Issue, (2010) (accepted for publication).

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1. A. K. Popov, S. A. Myslivets and V.M. Shalaev, "Nonlinear Coupling of Contra-propagating Electromagnetic Waves in Left-handed Nanocomposites," Progress In Electromagnetics Research Symposium PIERS 2009 in Moscow, August 18-21, 2009.

2. A. K. Popov, “Nonlinear optics of backward waves in negative-index plasmonic metamaterials,” 4th Rio de la Plata Workshop on Laser Dynamics and Nonlinear Photonics, Piriapolis, Uruguay, December 8-11, 2009.

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2. A. K. Popov and S. A. Myslivets, "Numerical Simulations of Negative-Index Nanocomposite and Backward-Wave Photonic Microdevices," Proc. of ICMS 2010: "International Conference on Modeling and Simulation," Cape Town, South Africa, January 29-31, 2010, Part 2, pp. 449-463, ISSN 2070-3724.

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1. Alexander K. Popov and Thomas F. George, ``Computational Studies of Tailored Negative-Index Metamaterials and Microdevices," Chapter 13 (47 pp.) to appear in Computational Studies of New Materials II: From Ultrafast Processes and Nanostructures to Optoelectronics, Energy Storage and Nanomedicine, Edited by T. F. George, D. Jelski, R. R. Letfullin, and G. Zhang, World Scientific, Singapore, 2010.

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Names of Post Doctorates

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This section only applies to graduating undergraduates supported by this agreement in this reporting period

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Compensating losses in optical negative-index materials and new frontiers in nanophotonics (final progress report)

Statement of the problem studied

Optical negative-index metamaterials (NIMs) present a novel class of electromagnetic materials that promise revolutionary breakthrough in photonics. Particularly, this regards with the signal and information processing capabilities and novel concepts of elemental and integrated optical components and devices, which enable smart, adaptive and reconfigurable sensing and image processing. The majority of NIMs realized to date consist of metal-dielectric nanostructures that have highly controllable magnetic and dielectric responses. Significant progress has been achieved recently in the design of bulk, multilayered, negative-index, plasmonic slabs (see, e.g., [1, 2]). The problem, however, is that these structures introduce strong losses inherent to metals that are difficult to avoid, especially in the visible range of frequencies and in the vicinity of negative-index resonance. Irrespective of their origin, losses constitute a major hurdle to the practical realization of the unique optical applications of these structures. Therefore, it is generally recognized that developing of efficient loss-compensating techniques is of a paramount importance. So far, the most common approach to compensating losses in NIMs has been associated with the possibility to embed amplifying centers in the host matrix. The amplification is supposed to be provided through a population inversion between the energy levels of the embedded centers. For the first time, such option has been successfully realized only few months ago [3].

OUR EFFORT seems to remain the sole one that explores an alternative option which offers compensating losses in double-domain positive/negative index (PI/NI) plasmonic nanocomposites without the requirements of population inversion or even without the requirements of doping. The approach proposed and developed in this research relies on direct coherent nonlinear-optical (NLO) energy transfer from the control optical field(s) to the negative-phase signal through three- (TWM) and four-wave mixing (FWM) propagation processes. The feasibilities of tailoring the transparency of the NIM slabs from strongly opaque via fully transparent to the amplifying states by the control laser(s) as well as the possibilities of unparallel applications of the discovered unique propagation properties have been demonstrated with numerical experiments. First optical NIMs were created in 2005; and nonlinear optics in the NIMs still remains the less developed area of electromagnetism. Contra-directed (negative) phase velocity and the energy flow (backwardness of electromagnetic waves) is intrinsic property of NIMs that never occurs in natural materials. It gives rise to counterintuitive electromagnetic phenomena, which require the developments of unparallel theoretical approaches and a revision of nonlinear propagation effects known from the textbooks on nonlinear optics. Hence, the results of this research and its outcomes, such as the proposed means of all-optically manipulating the transmission properties of plasmonic metamaterial slabs without changing their composition and structures, of energy transfer between the contra-propagating optical electromagnetic waves and manipulating their energy distribution across the slabs go far beyond merely the means of compensating losses in negative-index metamaterials. Overall, the major objective of this effort has been to investigate novel opportunities that could lead to a major breakthrough in photonics. This meets the goal of the Army Research Office to develop forefront concepts and approaches that would significantly improve information processing capabilities for the Army in the coming decades.

Summary of the most important results.

For the given metamaterial slab, negative index exists only within a certain wavelength band. Outside this band, refractive index is positive. A typical NLO coupling schemes proposed and investigated in this project include one negative-phase electromagnetic wave and two or three ordinary waves. In order to ensure phase matching, their phase velocities (wave vectors) must be co-directed. Hence, due to the backwardness of the negative-index wave, its energy flow appears counter-directed relative to those of all other, ordinary waves. Consequently, the set of the coupled Maxwell's NLO equations must be modified. In addition, the boundary conditions for one of the coupled waves must be applied to the opposite edge of the NIM slab. The coupling geometries may also differ depending on whether the input weak wave to be controlled falls in NI or PI wavelength range. These lead to fundamental differences in the solution procedure and, ultimately, to unusual solutions to the wave equations as compared with those known from the textbooks on nonlinear optics. The outlined issues have been addressed in our research. It appeared possible to present the equations for slowly varying amplitudes of the backward wave and for its ordinary coupled counterpart propagating in opposite directions in the form that can be used for either electric or magnetic, TWM or FWM nonlinearities provided that the control field(s) is fairly uniform across the NIM slab. The thickness of the currently available negative-index metamaterial slabs does not exceed the micrometer scale which is shorter than the lengths of picoseconds pulses. Therefore, the model of cw field has been employed since it suffices the investigations of the major problems set for the given effort. Specific features that would be attributed to very short, femto- and atto-second pulses propagating in a NIM slab, such as slow light, seems less actual at this point. Two options have been explored. One concerns the possibilities associated with the quadratic TWM nonlinearity intrinsic to the building blocks of the NIMs. The other one is aimed at independent engineering of resonantly enhanced cubic FWM nonlinearities and negative refractive index. FWM nonlinearities can be introduced by doping the host NIM with the resonant centers like ions, molecules or quantum dots. Due to multi-parameter dependencies, the investigations of each option require different theoretical approaches supported by the extensive numerical experiments with realistic models. Such experiments are instrumental for understanding the outcomes which often appear counter-intuitive.

First investigated option of the TWM based optical parametric amplification (OPA) of the negative-phase signal corresponds to the model where the dependence of the local optical and NLO parameters on the intensity of the control field and on the frequencies of the coupled waves can be neglected. In stark contrast with the slabs of ordinary materials, the solution to the equations for the coupled electromagnetic waves in this case shows that the intensities of the signal and the idler appear oscillating across the NIM slab even when the precise phase matching is fulfilled. Maximum amplitude inside the slab is typically much greater than the amplitude of the output signal unless a set of requirements that we have discovered is fulfilled. The amplitude of the output waves experiences an extraordinary resonance dependence on the product of the slab thickness and the control field intensity. Usually, such "geometrical" resonances are narrow and the slab remains opaque outside the resonances. Such behavior is in a strict contrast with the exponential dependence known for the counterpart OPA process in the ordinary NLO materials. The indicated resonance behavior may find various applications, e.g., for narrowband, frequency tunable filtering. Otherwise, it imposes the requirement of fine tuning of the intensity of the control field and therefore affects the robustness of transparency and amplification. The main important finding to report is that the extraordinary dependence of transmission of the negative-index backward-wave signal have been revealed on the ratio of its absorption coefficient and the absorption coefficient for the positive index idler. Counter-intuitively, it has been shown that transparency becomes robust and achievable within a broad range of these parameters at the expense of relatively small increase of the minimum required intensity of the control fields if the absorption for the idler exceeds that for the signal. This result was highlighted by peers (Nature Photonics, 3, 75, Feb. 2009, section "Metamaterials, news and views"). With the aid of numerical simulations, we have shown that nearly 100% transparency or even amplification of the negative-phase signal within a broad range of intensities of the control fields and the slab's thicknesses is possible. Thus, it has been demonstrated that the minimum transparency can be transformed, increased and even turned into amplification, so that it would remain robust for any magnitudes of the slab thickness and the control field intensity above a certain threshold.

Interesting phenomenon of tailored NLO reflection has been discovered and all-optically (remote) controlled ultra-miniature NLO mirror has been proposed and investigated. The phenomenon is based on the process of a difference-frequency generation of backward wave in a strongly absorbing double domain NI/PI slab which is related but distinct from the process of optical parametric amplification. In this case, incident strong control field produces tailored reflectivity for co-propagating incident PI wave which is accompanied by the frequency shift. It is shown that the reflectivity may significantly exceed 100% (amplified reflection) and the frequency of the reflected beam can be tailored within the negative-index frequency domain.

On a fundamental level, NLO response of the nanostructured metamaterials still remains little understood, characterized and cannot be predicted effectively so far. However, it is well established that the nonlinearities attributed to the plasmonic nanostructures can be strongly enhanced by the local fields. Only rough estimates of their magnitude are available to date. Assuming quadratic nonlinearity on the order of that characteristic for the best nonlinear-optical crystals, our estimates show that with the control field intensity on the order of 100 kW focused on the spot of the diameter about 50 μm the required metaslab thickness is estimated on the order of few μm.

With account for the aforementioned problems with the plamonic nonlinearities, part of our research thrust was focused on investigating the possibilities of independent engineering of NLO response and the NI by embedding resonant NLO centers in the host NIM. Several models have been investigated for which strong FWM NLO response of the composite is primarily determined by the embedded resonant four-level centers. Hence, such nonlinearity can be engineered and adjusted independently. In addition, the feasibility for quantum control of the intensity-dependent local linear and NLO parameters has been shown that employs constructive and destructive quantum interference tailored by two driving control fields at the transitions between the energy levels of the embedded centers. Original density-matrix theory has been developed that enables one to include empirical relaxation parameters which crucially determine the indicated intensity dependence of the local parameters. Such account for the variety of different important relaxation parameters is not possible in the framework of the Schrödinger equations. The method developed allows one to obtain an analytical solution for the intensity-dependent local optical parameters. Ultimate solutions for the transmission factor and for the distribution of the signal and the idler across the NIM slab exhibit strong multi-parameter dependences that can be analyze and the maximum output can be found only through numerical experiments.

The theory of resonant four-wave mixing (FWM) of ordinary and backward waves has been developed. Contrary to the off-resonant processes, here, real and imaginary parts of the linear and nonlinear susceptibilities play equally important role. Numerical models with different relaxation parameters and different intensities and resonance frequency offsets for the control fields have been employed in order to understand multi-parameter dependences of the extraordinary processes under the investigation and to find the characteristic optimal parameters for the doping agent and for the control fields. In the first period of our research, it seemed obvious that Raman-like or population-inversion gain of the ordinary, positive index idler would greatly support the direct coherent energy transfer from the PI control field to the NI backward-wave signal and thus benefit the transparency. However, our research during the reporting period has shown that there exists an alternative option which leads to the different class of schemes for engineering the embedded nonlinearity and has important applications. As indicated above, it was found out that the transparency of a NIM slab becomes essentially more robust against the intensity of the control field and the phase mismatch provided that absorption index for the PI idler is larger than that for the NI signal. Based on this outcome, the studies of the resonant NLO coupling and the accompanying quantum interference processes have been carried out for the alternative option with regard to the doping agents that assumed unfavorable and had not been explored earlier. Here, the signal appears in the vicinity of the transition between the excited energy levels, whereas the higher-frequency idler couples with the ground state of the embedded quasiresonant centers so that such a scheme provides a different type of the resonantly enhanced FWM response. As indicated, resonant and quasi-resonant coupling and the output NI signal can be tailored through the means of quantum control. It appeared a complex multi-parameter optimization problem that had required extensive numerical experiments on the local parameters driven by the control fields, on the distributions of the contra-propagating signal and idler across the NIM slab, and on their output values. In the given scheme, all the dependencies are different from those in the alternative earlier investigated scheme. A model with a set of the relaxation parameters has been chosen which does not allow population inversion or Raman gain at any of the involved resonant transitions. Ultimately, nonlinear propagation for such a model was monitored through the numerical simulations. It has been shown that the outlined cardinal changes in the coupling scheme bring about major changes in the properties of the laser-induced transparency of the doped NIM slabs. The challenge is that we propose to achieve transparency for a strongly absorbing host material by doping it with the impurities that introduce additional strong resonance absorption. The numerical simulations have proven the feasibility of tailored transparency and amplification for the NI signal in such coupling schemes. Ultimately, the earlier investigated schemes with amplification for the idler provides for easier achievable oscillation threshold and narrow-band amplification, whereas the described alternative option offers more robust and potentially more broad-band transparency while no incoherent amplification at any of the involved transitions is required. Changes in the relaxation and spectral properties of molecules embedded in the metal-dielectric nanocomposites, e.g., in the fishnet nanostructures are little known and not completely characterized to-date either. So we could rely only on the reasonable assumption for our numerical models. In such context, our research presents the attempt to look ahead and to answer the basic questions of whether compensating losses in the NIMs through direct coherent energy transfer from control ordinary fields to the backward signal is possible in the situations where neither population-inversion nor Raman gain are achievable and, if so, what are the typical requirements and whether they are practical. Among the important outcomes of this part of the effort is the feasibility of improving the phase-matching through a proper adjustment of imaginary and real parts of the FWM susceptibilities by the adjustment of the frequency resonance offsets for the control fields. Such option is generic to quasi-resonant nonlinear optics but not available in the ordinary, far-off-resonant nonlinear optics. The extensive numerical experiments have shown that the required intensities of the control fields fall within the range of one Watt to several kW per a cross-section of several parts of mm, depending on the frequency resonance offsets and on the relaxation parameters of the quantum system. At number density of the impurities on the order of 10^19 cm^-3, the required slab thickness is estimated on the order of few microns. We expect that utilization of the impurities with much lower coherence relaxation rate than in molecules, e.g. spin coherence in rear-earth ions, would make laser-induced transparency even easier achievable, whereas the operational wavelengths fall in the telecommunication band.

As outlined, besides compensating losses in strongly absorbing NIMs, the discovered extraordinary propagation features open an avenue for creation of novel photonic components and devices. Among them is novel class of the miniature frequency-tunable narrow-band photonic filters, quantum switches, tailored nonlinear-optical micromirrors, amplifiers and cavity-free microscopic optical parametric oscillators (OPOs). The unique features of the proposed photonic devices are revealed, such as described strongly resonant behavior with respect to the material thickness, the density of the embedded resonant centers and the intensities of the control fields. The possibilities of narrow-band frequency-tunable switching and filtering, as well as of the backward-wave mirrorless optical parametric oscillations (BWMOPO) in the microscopic samples of NIMs have been proved through numerical experiments. BWMOPO enables generation of counter-propagating entangled right- and left- handed photons. Note, that three-wave parametric coupling in ordinary transparent crystals for the signal and the idler with energy flows and wave-vectors co-directed for each wave but anti-parallel for the different waves was proposed in the beginning of the NLO era by the several groups of authors. The possibility of extraordinary mirrorless self-oscillations was predicted. Particularly, such a possibility was described by S. Harris in his paper published in Appl. Phys. Lett. in 1966. The results appeared so unusual and remarkable that they were included in the textbook on quantum electronics by A. Yariv in 1973 as a curious example despite the fact that phase-matching in such a case seemed practically impossible. It would require anomalous dispersion and close frequencies of the pump and one of the generated waves so that the other one would appear in the far infrared range were crystals are strongly absorptive. For the first time, such OPO was realized 41 years later, in 2007, by employing the nanostructured periodically poled crystal which became possible owing to recent advances in nanotechnology. This breakthrough was marked as the achievement of the year at topical OSA and APS meetings. Our proposal is different, because all wave vectors are parallel whereas energy flows for the signal and the idler appear counter-directed. Such a coupling scheme is natural to NIMs. It removes a formidable phase-matching obstacle which is fundamental for the ordinary crystals and opens an avenue for realization of extraordinary features associated with NLO of backward electromagnetic waves and for their applications.

Most of the numerical experiments were performed for the models that assume homogeneous distribution of strong control fields across the slab. It was shown that absorption of the control field causes some quantitative changes, whereas basic qualitative features remain unchanged. Considerable computation problems have been encountered at investigating the regimes that are accompanied by the significant depletion of the control fields due to their conversion to the signal and idler. Solutions found for the specific ultimate cases shows substantial qualitative changes that require further investigations.

The reported results have been presented at major topical international conferences (including invited talks) and published in the peer reviewed journals [4-14]. The basic ideas regarding optical properties of nanoparticles and nanostructures, the extraordinary electromagnetic properties of NIMs, as well as the related revolutionary changes in electromagnetics and in optics beyond the principles described in the textbooks were presented at the meeting of the instructors in physics, astronomy and computer science at the University of Wisconsin Colleges. Selected topics related with the described ideas were included in the undergraduate course on general physics offered to students of the University of Wisconsin-Fox Valley. Several relevant problems in mechanics, electricity, magnetism and optics for undergraduate students were created and offered to the UW-Fox Valley students and discussed at the meeting with the instructors. Follow-up questions and e-mails from students expressed their interest in further learning in nanoscience. A chapter on basic principles of nanophotonics has been written and included in the textbook for undergraduate students [15]. Unfortunately, no students were available for the research, primarily because the PI holds non-teaching position sponsored by this project.

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8. A. K. Popov, S. A. Myslivets and V.M. Shalaev, ``Plasmonics: nonlinear optics, negative phase and transformable transparency (Invited Paper), “ in Plasmonics: Nanoimaging, Nanofabrication, and their Applications V, edited by Satoshi Kawata, Vladimir M. Shalaev, Din Ping Tsai, Proc. of SPIE Vol. 7395, 73950Z-1(12) (2009).
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